Time-Dependent Studies of Photoionization of Light Systems: Beyond Two-Electron Systems

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In the last 10–15 years, significant progress has been made in solving, by a variety of powerful theoretical methods, the Coulomb three-body problem inherent in the double photoionization of a two-electron atom (γ,2e), or in the electron-impact ionization of a one-electron system (e,2e). These problems are found in their most basic form in the double photoionization of helium and the electron-impact ionization of hydrogen. Recently, as many as four distinct methods have found great success when applied to the full solution of these problems. These include the convergent close-coupling [1], the exterior complex-scaling [2], the hyperspherical R-matrix with semiclassical outgoing waves [3], and the time-dependent close-coupling methods [4]. Total, single, double, and triple differential cross sections have been calculated for these systems for a variety of different dynamical situations and, in general, very good agreement has been found with recent experimental measurements of these quantities.

It has therefore become natural to consider problems beyond this level of complexity. In particular, Coulomb four-body problems have recently been of interest due to experimental measurements of the triple photoionization of lithium and many measurements of cross sections associated with the double photoionization of the hydrogen molecule, H_2. The triple photoionization of lithium involves three charged particles moving away from a charged nucleus, and the double photoionization of H_2 entails two electrons being ejected from a diatomic molecule (which itself can then fragment).
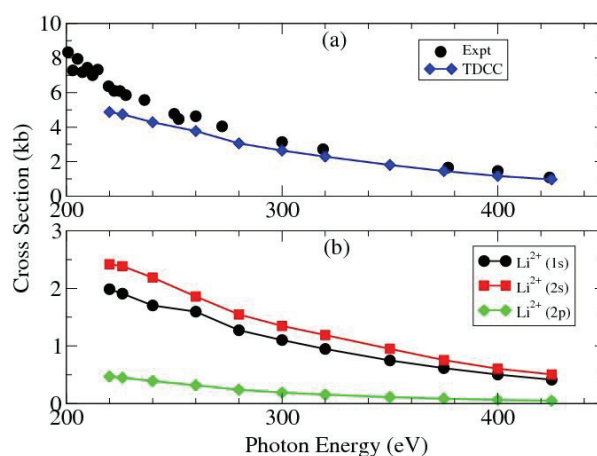
Recently, the time-dependent close-coupling technique has been used to numerically investigate these problems. The time-dependent method was extended to treat equally three outgoing electrons moving in the field of a nucleus by propagating in time a 9D wave function according to the Schrödinger equation [5]. The three radial dimensions were represented on a lattice and a coupled channels expansion represented the remaining six angular dimensions. After propagation in time, double photoionization (with excitation) and triple photoionization cross sections for lithium were obtained by projection onto fully antisymmetric spatial and spin functions. In Fig. 1, we show on the left the radial part of the ground state of lithium on a 3D lattice. This ground state represents the starting point for the real time propagation. On the right we show the double photoionization cross sections as compared with recent experiments. The agreement with experiment is very good and we are also able to distinguish in which final state the Li^{2+} ion is left. It is hoped that planned experiments will also be able to measure these double photoionization (with excitation) cross sections. Triple photoionization cross sections have also been calculated and are in good agreement with other experimental measurements.

The time-dependent calculations as applied to molecular systems started with a preliminary investigation of single- and multi-photoionization of the one-electron system H_2^+ [6]. These calculations were made in an effort to test our new computer programs against previous theoretical work. In this case, the radial and one angular dimension of the electron were represented on a lattice for each value of the remaining angular dimension. For the time being, the internuclear separation is held fixed in these calculations. The single-, two- and three-photon ionization rates were found to be in good agreement with previous calculations made using time-independent techniques.
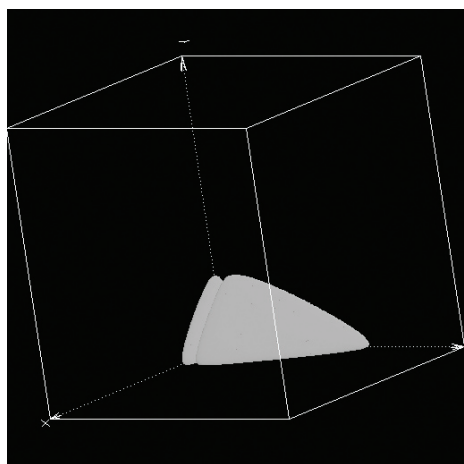
This work allowed us then to pursue calculations on the two-electron H_2 system [7]. Double photoionization calculations of H_2 were now made by representing two radial and two angular dimensions on a lattice and propagating in time this coupled wave function according to the Schrödinger equation. As before, photoionization cross sections are obtained by projecting onto suitably antisymmetrized functions. In this case, the double photoionization cross sections were in good agreement with experimental measurements. We now are applying our methods to look at angular differential cross sections, as well as examining the role of nuclear motion in these molecular calculations.

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